

Substantial Enrichment of Mechanical, Thermal and Electrical Properties of Thermoplastic Polyester Elastomer by Melt Blending with Nano-ZnO

Haydar U. Zaman^{1,*}, Ruhul A. Khan²

Abstract

Polymers are more reinforced with nano metal oxides, a completely flexible composite of engineering that simplifies the way of exhibiting good mechanical and chemical features. Polymer mixtures represent a very vital field in the handling of innovative ingredients, which have better features than net polymers. Polymer mixtures are able to deliver ingredients with prolonged useful features beyond the range that can be achieved from a single polymer equivalent. A methodical study was performed to test the features of the matrix by introducing nano-ZnO (4 nm, 1–5 wt%) into the matrix from a poly(butylene terephthalate)-block-poly(tetramethylene glycol) (PBT-PTMG)-based thermoplastic polyester elastomer (TPE). ZnO nanoparticles were coated with maleated styrene ethylene butylene styrene (SEBSMA) before melt blending for better surface bond and fine dispersion. The effects of uncoated and coated nano-ZnO (nZnO) particles with varying concentrations on the mechanical, thermal and electrical features of binary TPE/nZnO nanocomposites were manufactured by the melt compounding process tracked by hot press mold. The tensile features such as yield strength, tensile strength, tensile modulus, and elongation at break varied with the concentration of nZnO. The mechanical features exhibited that the strength and modulus increased with increasing nZnO loadings while the elongation at break recorded a linear decrease. The morphological observation revealed that the filler dispersed well in the polymer matrix due to the formation of chemical bonds. Nano-ZnO incorporation enhances electrical and thermal features such as melting temperature, and thermal stability of nanocomposites.

Keywords: Thermoplastic polyester elastomer, nano-ZnO, nanocomposites, mechanical and thermal properties

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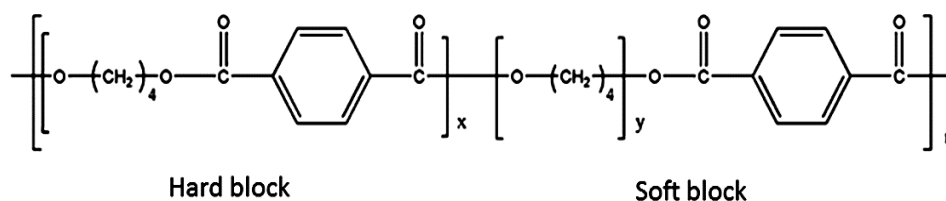
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INTRODUCTION

Thermoplastic elastomers (TEs) are increasing in importance as inventive materials, which cover the boundary among rubbers and plastics. TEs consist of a thermoplastic polyester elastomer (TPE), consisting of poly(butylene terephthalate) (PBT) as a hard section and poly(tetramethylene ether glycol terephthalate) as a soft section, with superior mechanical, thermal features, and elasticity [1]. TEs are the materials, which demonstrate their unique strength, flexibility, and processing that occurs due to the individual microstructures in their step. When elastomers are processed at a high temperature the solid phase melts and evaporates. The properties of TE depend on the nature and amount of hardening phase in the copolymers. Due to its great processing and elasticity, TE is widely used in various fields such as the automotive industry, electrical equipment,

sports, etc. [2]. To date, various TEs such as styrene, urethane, amide, olefin, and ester have been commercialized, providing their applications for automotive and home appliances [3–6]. Thermoplastic polyester elastomers (TPEs) in TEs have more outstanding properties, for thermal and mechanical performance [7]. A family of innovative segmented TPE has been flourished by du Pont under the trademark Hytrel®. TPE is a type of thermoplastic that is made up of a solid component of the block structure polybutylene terephthalate (PBT) and is classified as a soft section consisting of polyteramethylene glycol (PTMG). The chemical structure of PBT-PTMG based TPE has been revealed in Scheme 1.



Scheme 1. Chemical structure of the thermoplastic polyester elastomer (TPE).

Owing to significant processability and elasticity, superior oil and chemical resistance, superior lightweight, fatigue-resistance, recyclability, excellent heat resistance performance, alongside fantastic low-temperature impact strength, TPE is extensively used in the areas of automotive parts such as hoses, gears, tubing, and gears, and electronics, electronic parts, and even increasingly substitutes the cross-linked rubbers [8]. TE has an elastomeric performance at room temperature, which can evaporate at higher temperatures than common thermoplastic. Binary mixtures of polystyrene (PS) and poly(vinyl methyl ether, PVME) are miscible and several rumors have been expressed about the mechanism for interaction. J.H. Ryou et al reported [9] that there is an interaction between the ether group and aromatic ring of PS, based on a change in the IR spectrum of the miscible and immiscible PS/PVME film.

The agglomeration of inorganic nanoparticles in the polymer matrix can significantly affect the properties of the matrix [10]. Acquired composites may be rich in mechanical, thermal, or electrical properties. These hybrid materials are polymers with structural flexibility and high carrier mobility, bandgap tenability resulting in overlapping effects on their unique properties. Therefore, they can allow a versatile design of their physical and chemical features, meeting the needs of end-user applications [11]. The features of polymer nanocomposites depend on the type, size, and shape of the integrated nanoparticles, and interactions with the polymer matrix [12]. The key to polymer nanocomposite technology is the barrier to particle agglomeration. Several nanoparticles have been integrated into the polymer matrix for nanocomposite applications. One of the reasons inorganic nano-metal oxide (nZnO) is that nZnO is a cheap, less toxic property for diabetes treatment, wound healing, antibacterial, drug delivery, anti-cancer, anti-inflammatory, and high biomechanics, high biomechanics, stability large electronic machine coupling coefficient, high catalytic activity, high illuminated transmittance, intensive UV and infrared absorption, etc. [13–15]. The role of ZnO nanoparticles in polymers can lead to the development of mechanical and optical properties of polymers due to their small size and large specific fields and quantum effects of organic polymers and inorganic nanoparticles, respectively.

TPE/nZnO nanocomposites have attracted great research interest due to their flexibility in design, in addition to improved mechanical strength, better chemical, electrical and optical properties. Despite a substantial increase in mechanical properties, particles can signify a decrease in toughness due to their hardness. As a result, an excellent balance of stiffness to toughness can be achieved by incorporating both elastomeric phase and inorganic reinforcement into the polymer matrix. The most practiced elastomers are styrene-ethylene-butylene-styrene (SEBS) [16], ethylene-propylene-diene monomer (EPDM) [17], and ethylene-propylene rubber (EPR) [18] to increase the toughness of hybrid composites. One of the problems that can be encountered while studying nanoparticles is the agglomeration of particles Agglomerates in the matrix reduce the aristocracy of the composite and as a result, dispersants and compatibilizers must be used to overcome this problem. Styrene-ethylene-butylene-styrene grafted maleic anhydride (SEBS-g-MA) or maleated polypropylene (PPMA) is

commonly used in hybrid composites. Tjong et al. [16] used SEBS-g-MA in PP/montmorillonite nanocomposites. Whereas SEBS increased the toughness of the composite, maleic anhydride acted as a compatibilizer between the inorganic particles and the matrix. Several researchers have studied the physical and mechanical properties of ZnO with PP composites [19, 20]. In this study reported herein, the mechanical, thermal, and electrical properties of selected TPEs are influenced by the agglomeration of nano-ZnO particles and particle contents.

EXPERIMENTAL

Materials

The polymer matrix TPE granules were kindly delivered by M/S Rupal Plastics Ltd, Mumbai, India with a grade was Hytrel 6356. The weight percentage and molecular weight of the PTMG part in this PBT-PTMG copolymer are approximately 50% and 100 g/mole, respectively. The nano-ZnO particles used in this study with an average size of 20 nm were purchased from Changtai New Material Co., Ltd., China. Approximately 10% SEBSMA (Kraton FG-1901X), with 1.84 wt% MA graft ratio and MFI of 20 dg/min (270°C/5kg), obtained from Shell Chemical Co (Houston, TX) were used as a compatibilizer to modify particles.

Methods

Modification of ZnO Nanoparticles with Compatibilizer

Before modification, nZnO particles were dried under a vacuum oven. SEBSMA was used by dissolving toluene at room temperature for 48 h. Then, pure nZnO particles are gently included in this solution and mixed with a mechanical mixer (IKA RW 20 digital) for 2 h. The mixture was powdered after drying at 50°C for 8 h. The amount of the SEBSMA was 5% of the nZnO particle.

Nanocomposite Preparation

Before compounding, all the components were dried in a vacuum oven at 60 °C for 48h and then cooled down to room temperature before use. All ingredients were incorporated into the extruder immediately before mixing. The extrusion of a TPE was performed by totaling 1, 3, and 5 wt% of nZnO into a Berstorff twin-screw extruder (ZE-25A UTX, KraussMaff ei Berstorff GmbH, Germany). The coding of the nanocomposites was done as follows: TPE/1nZnO, TPE/3nZnO, and TPE/5nZnO. The diameter of the screws was 25 mm and the length/diameter ratio (L/D) was 44. Processing temperatures ranged from 200°C in the neighboring hopper to 220°C in the die and the speed of the screw was 150 rpm. The resultant extruded was cooled and ground into small pellets. The obtained pellets were used to make thin plates in the electrically heated hydraulic press. After 5 min of preheating, the pellets were hot-pressed for another 5 min at 210°C and then water-cooled to ambient temperature with a cooling rate of 10°C/min.

Characterizations

Surface Morphology and Particle Dispersion

SEM was used to analyze fracture surfaces and the dispersion of nanoparticles. SEM, JSM-6360LV from JEOL, Tokyo, Japan was used to analyze the fracture surfaces of nanocomposites. The samples were gold-coated before imaging.

Differential Scanning Calorimetry

The crystallization, melting behaviors, and crystalline enthalpy of nanocomposites were investigated by differential scanning calorimetry (DSC, Perkin Elmer DSC-7) in the N₂ atmosphere. Each sample (5–8 mg) was heated from 30 to 280°C at a heating rate of 10°C/min and then held at 280°C to confirm a duplicate thermal history. The specimen was cooled to 30°C at a cooling rate of 10°C/min.

Thermogravimetric Analysis

The thermal stability of the nanocomposites was determined using a thermogravimetric analyzer (TGA, TA Instruments Q500) with a heating rate of 20°C/min in an N₂ atmosphere.

Mechanical Properties Evaluation

The tensile test was measured using Shimadzu Universal Testing Machine (model AG-1, Japan). Samples were ~ 10 mm wide, 2 mm thick, with a gauge length of 50 mm. A crosshead speed of 10 mm/min was used to measure the yield strength, tensile strength, tensile modulus, and elongation at break. All experiments were conducted under ASTM-D 638-14, standard [21], and five repeat tests were performed to obtain an average value for each sample.

Electrical Tests

Uncoated and coated nanocomposites were cut into very small pieces and then powdered to make tablets. This tablet has been used to measure the dielectric properties. A Marconi Universal Bridge (TF 1313 A) is used to measure the dielectric properties at 10 kHz. The dielectric constant and resistivity of nanocomposites were measured using the bridge technique (two-electrode method) at a frequency of 10 kHz. An electrometer (model-617) was used for measuring resistivity.

RESULTS AND DISCUSSION

Dispersion of Nano-ZnO Particles in TPE Matrix

It is well-known that the dispersion of filler in the polymer matrix can have a noteworthy influence on the mechanical properties of the nanocomposites. Dispersion of a thermoplastic inorganic filler is not a simple procedure. The problem is exacerbated when used as a nanoparticle filler, as nanoparticles have a strong tendency to aggregate. Figure 1 (a–c) represents the TEM photomicrographs of TPE/3

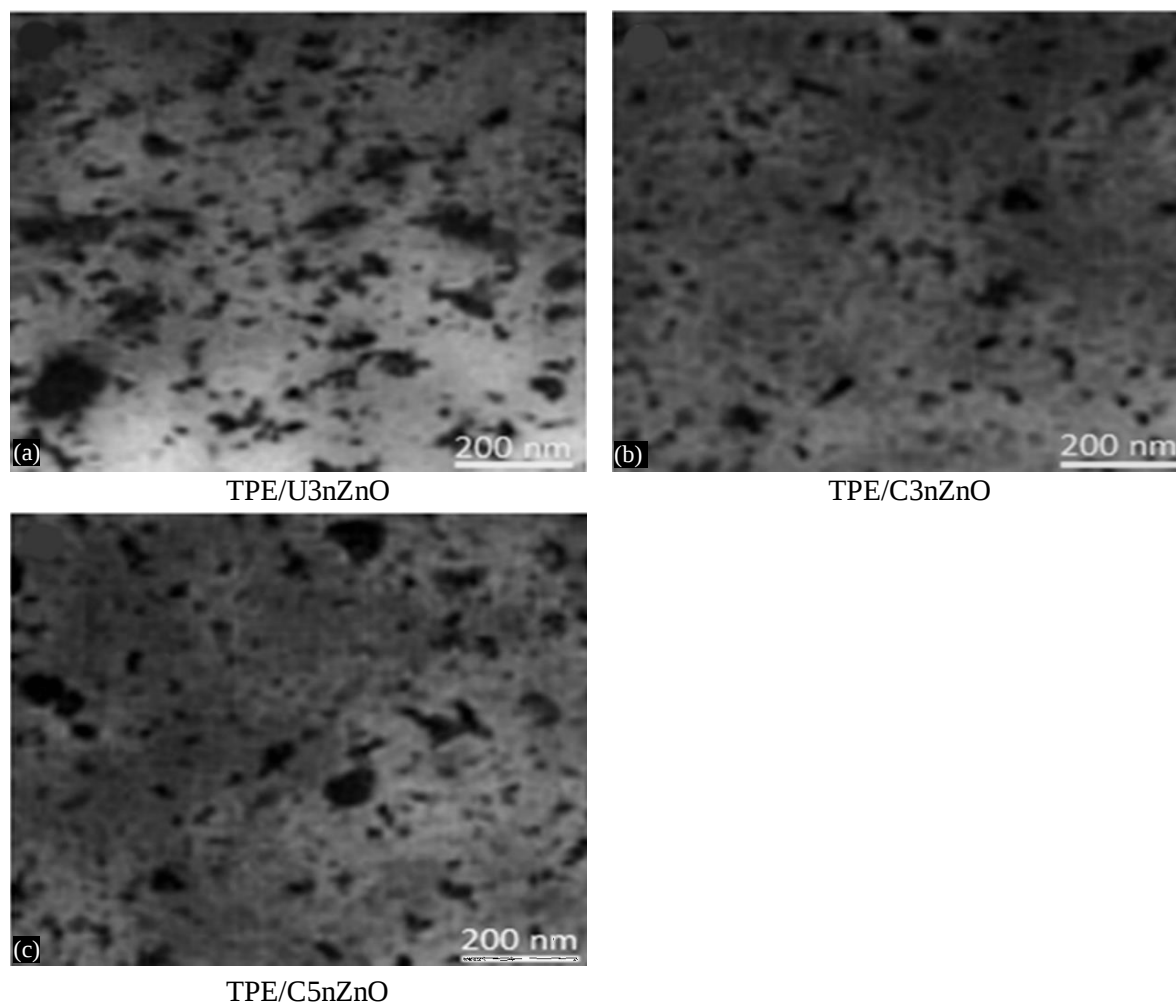
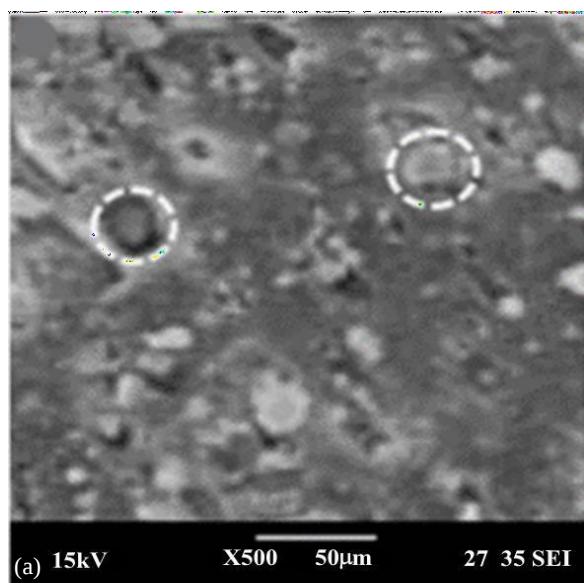


Figure 1. (a–c) TEM photomicrographs of TPE/nZnO nanocomposites (w/w%) (a) TPE/U3nZnO, (b) TPE/C3nZnO, (c) TPE/C5nZnO.

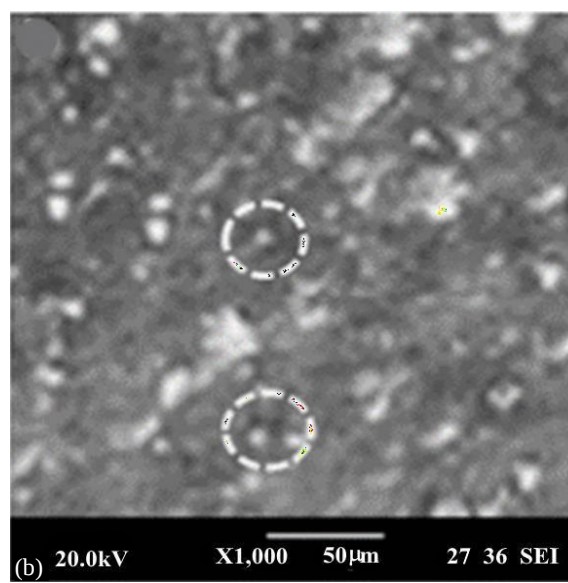
wt% uncoated nZnO (3UnZnO), TPE/3 wt% coated nZnO (3CnZnO) and TPE/5 wt% coated nZnO (5CnZnO) nanocomposites, respectively. In the TEM image, the dark regions are ZnO nanoparticles. In the TEM image (Figure 1, b) most nZnO particles are uniformly dispersed in the TPE/3CnZnO nanocomposite. TEM photomicrographs showed that most nanoparticles are less than 100 nm in size and the smallest are less than 5 nm.

Morphological Characterization of Nanocomposites

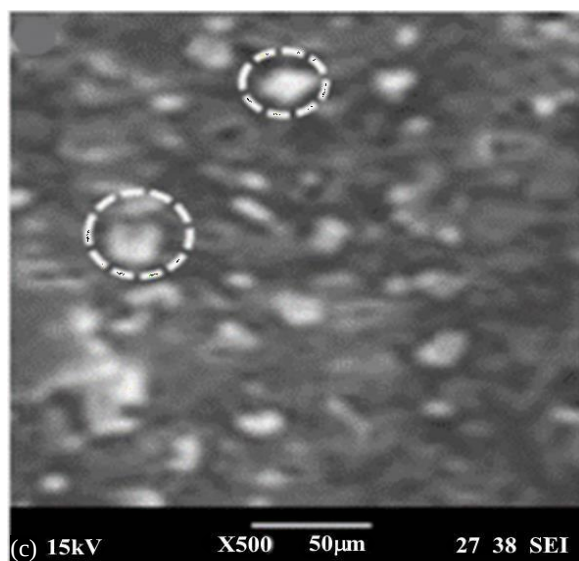
Mechanical properties are related to the uniformity of multiple morphologies, domain sizes, and shapes to determine the degree of dispersion of interactions between the two stages, and it is considered to be one of the most important elements [22]. Generally speaking, an operative compatibilizer gives polyblend with rich phase domain size, better interface contact area, and upper interfacial bond. The morphological properties of TPE/nZnO nanocomposites were examined using SEM. Figure 1 (a-c) demonstrates the morphological evidence of fractured surfaces of TPE/U3nZnO, TPE/C3nZnO, and TPE/C5nZnO nanocomposite samples. Figure 2(a) displays that the nZnO particles dispersed randomly in TPE/U3nZnO and that several large agglomerates with particle sizes greater than 1 μm were present on the fracture surface, which indicates poor compatibility between TPE and nZnO. nZnO particles were dispersed unevenly in the TPE and lots of particles were agglomerated and conducted into larger cavities around the agglomerates. It makes sense that hydrophilic ZnO nanoparticles are agglomerated into TPE. A specific quantity of hydroxyl groups can form hydrogen bonds between nanoparticles and TPE, as a result, higher agglomeration occurs. Larger nanoparticles are dispersed in TPE/nZnO nanocomposite so that there are no functional polymers and thus the interfaces are individually wetted and/or the bonding of the components appears to be somewhat weak. This allows for the lower tensile properties of the nano-composites discussed later. Realizing this agglomeration, we found that these were formed by many elementary particles. Figure 2(b-c) displays SEM images of TPE/C3nZnO, and TPE/C5nZnO nanocomposites, respectively. The presence of TPE/3nZnO or TPE/5nZnO changes the morphology. The fractured surface morphology of the TPE/C3nZnO nanocomposite was more uniform and finer than that of TPE/C5nZnO nanocomposite, and the mean particle size was smaller than that of TPE/C5nZnO nanocomposite. Once the nano-ZnO surface was coated with SEBSMA, it randomly distributed into TPE, filling cavities, and the interfacial interaction between TPE and nano-ZnO was enhanced by the effect of SEBSMA. Also, the inclusion of C3nZnO in the TPE matrix was more homogeneous, and the cohesive force between them was stronger. SEBSMA has covered nZnO and improved compatibility between the TPE matrix and nZnO. These satisfactory nanoparticles and holes can persuade large-scale plastic distortion of the TPE, which increases the strength of the fracture.



TPE/U3nZnO



TPE/C3nZnO



TPE/C5nZnO

Figure 2. (a–c) SEM images of TPE/nZnO nanocomposites (w/w%) (a) TPE/U3nZnO, (b) TPE/C3nZnO, (c) TPE/C5nZnO.

Thermal Characterization

The DSC outcomes of virgin TPE and its nanocomposites (TPE/nZnO) are displayed in Table 1. From the outcomes, the addition of ZnO nanoparticles to TPE displayed a potential effect on the melting temperature (T_m) of the PTMG segment. At 1 wt% of ZnO nanoparticles, the T_m of the PTMG segment increases from 88.3°C of the matrix to 99.5°C of nanocomposites. At 5 wt% of ZnO nanoparticles, the T_m of the PTMG segment in nanocomposite was 5.1°C lower than that of virgin TPE. In contrast to the severely affected T_m of the PTMG segment in nanocomposites, the T_m of the PBT segment of the TPE/nZnO nanocomposite is only slightly increased compared to the TPE matrix, yet it exhibits a similar decreasing tendency with nanoparticle content. The highest enlargement in T_m (PBT) is 4.5°C occurring at 1 wt% of ZnO nanoparticles. The growth of T_m of PTMG and PBT segments in nanocomposites should be approved for chemical bonding between matrix and ZnO nanoparticles. In contrast, the inclusion of ZnO nanoparticles in TPE can reduce the weight fraction of PTMG and PBT segments in the composition. This may be due to a decrease in T_m (PTMG) and T_m (PBT) in nanocomposites with increasing nanoparticle content. Moreover, it was believed that most of the chemical interactions between the matrix and ZnO nanoparticles are performed on the surviving nanoparticles in PBT/PTMG amorphous phase in which the PTMG segment is enormous. As a result, the increasing effect on melting temperature is more pronounced for the PTMG section than for the PBT section. As an alternative result of chemical bonding between the matrix and ZnO nanoparticles, the crystallization of the hard PBT section in nanocomposites decreased due to the reduction of the chemical regularity of the macromolecular chain, characterized by the reduction of the crystal enthalpy (ΔH_c) presented in Table 1. From Table 1, the crystalline temperature (T_c) of the PBT section in the nanocomposite is transferred to a higher temperature. This increase in the T_c of the PBT section in nanocomposites should be recognized for the nuclear effect of ZnO nanoparticles. The list of the entire datum obtained in DSC examinations is given in Table 1.

Table 1. Thermal properties of virgin TPE and TPE/nZnO nanocomposites.

NCs (wt%)	T_m (°C)	T_c (°C)	ΔH_c (J/g)	T_{onset} (°C) (TGA)
	PTMG block	PBT block	PBT block	
V. TPE	88.3	176.8	134.6	25.2
U3nZn	99.5	181.3	136.5	22.1
C3nZnO	96.2	178.6	137.3	21.9
C5nZnO	94.4	177.5	136.9	21.4

NCs: Nanocomposites; V: Virgin; U3nZnO: TPE/U3nZnO; C3nZnO: TPE/C3nZnO

The TGA investigation conveys significant evidence in determining the thermal stabilities of the nanocomposites. Thermal stability is a significant property for which the nanocomposite morphology played a vital role [23]. The TGA thermograms of virgin TPE and TPE/nZnO nanocomposites are displayed in Figure 3. It can be assumed that all the samples exhibit a one-step degradation process in the temperature range of 349–470°C. The weight loss of virgin TPE and its nanocomposites behind 349°C is less than 0.02% with remarkable moisture loss in the matrix. Key decomposition related to matrix backbone reduction in virgin TPE starts at about 349°C with an acute rate. The onset degradation temperature (T_{onset}) of TPE is 393.5°C, which indicates the thermal stability of polymer material. The inclusion of ZnO nanoparticles strengthens the thermal stability of the matrix, characterized by an increase in the thermal degradation temperature of nanocomposite with increased nanoparticles content. The maximum increase in thermal stability is 8.9°C at 3 wt% of nanoparticles.

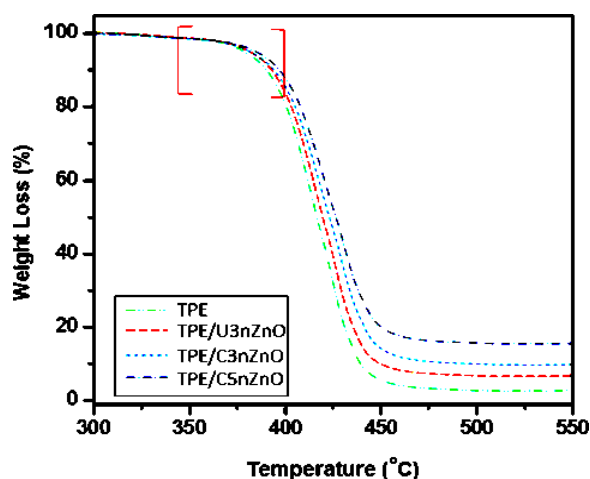


Figure 3. TGA curves of TPE and TPE/nZnO nanocomposites.

Mechanical Properties of the TPE/SEBS Blends

Metal oxide nanoparticles progress the mechanical features of the polymer matrix [24]. Figure 4 displays the variation of yield strength and tensile strength of TPE, TPE/UnZnO nanocomposites with nZnO contents varying from 0 to 5 wt% and TPE/CnZnO nanocomposites with the same contents against nZnO. It can be observed that increasing the nZnO content up to 3 wt% increased the yield strength and tensile strength (yield strength increased by 20% and tensile strength increased by 25% for TPE/U3nZnO nanocomposites and yield strength increased by 43% and tensile strength increased by 38% for TPE/C3nZnO nanocomposites compared to TPE, respectively) and then decreased by 5 wt%. Increased yield and tensile strength are recognized by mixing the nanofiller spread with the increased features. The addition of nZnO particles to the TPE improves the yield and tensile strength of the matrix at 3 wt% nZnO due to strong stress transfer from TPE to nZnO. At 5 wt% of nZnO particles, an improvement in yield and tensile strength was reduced due to the relative low-stress transfer effect of nZnO particles. In this case, the agglomerated nanoparticles are easily deboned from the TPE and no fraction of the external load is ultimately allowed to reduce the yield and tensile strength. These outcomes were supported by Zaman et al. [25]. To improve the interfacial bonding between nZnO particles and TPE, SEBSMA led to the modification of TPE/nZnO nanocomposites. The addition of SEBSMA in TPE/nZnO blends has created significantly noticeable interfacial bonding between filler and matrix than those of TPE/nZnO nanocomposites. The maximum yield and tensile strength of the TPE/C3nZnO nanocomposite were 23.9 MPa and 25.3 MPa at 3 wt% of nZnO content, respectively, and about 43% and 38% higher than that the TPE matrix. The increase in the strength of the compatibilized system is expressed by the better distribution produced by the compatibilizer and an enhanced solid-state adherence, which can transfer more stress from the matrix to the dispersion phase. MA functional groups have been grafted into the ethylene butylene mid-block of SEBS. Then, it converts to TPE from SEBSMA molecular chains and bonds with metal oxides. Consequently, homogenous, fine dispersed structures can be achieved.

Figure 5 shows the variation of tensile modulus and elongation of TPE/nZnO nanocomposites against nZnO content. With the addition of nZnO, tensile modulus steadily increased while elongation at break steadily reduced. Modulus growth offers an effective stress transfer from the TPE to nZnO particles. On the other hand, the elongation reduced with the addition of nZnO indicated interference or deformability of TPE by nZnO. This interference was created by the presence of physical interaction and mechanical restraint of the TPE. Maximum TM (303.6 MPa) of TPE/UnZnO nanocomposite was obtained at 5 wt% of nZnO content. At this stage, the TM value of the nanocomposite was about 48% higher than that of the TPE matrix. According to the trend of the modulus variant, the TPE/CnZnO nanocomposite increased more evidently than TPE/UnZnO nanocomposite. Consequently, the elongation at break was reduced with nZnO content indicating intervention by nZnO in the mobility or deformability of the TPE. This intervention was created by the physical interaction of TPE and the presence of mechanical restraint. As the concentration of nZnO increases, the elongation at break decreases at intervals, which describes an increase in the rigidity of the TPE nanocomposite. However, SEBSMA coated nZnO nanocomposites showed lower elongation at break than uncoated nZnO nanocomposite. These results agree with morphological observations and provide a strong interaction between nZnO and TPE, which is more important than TPE because TPE has modified nZnO due to better expansion and facial stress bridge from TPE to the nZnO.

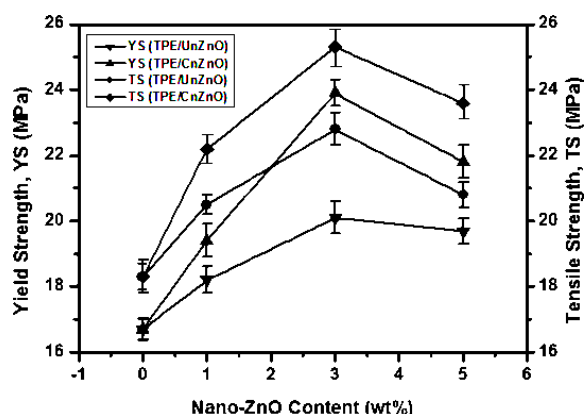


Figure 4. Yield strength and tensile strength of TPE/nZnO nanocomposites.

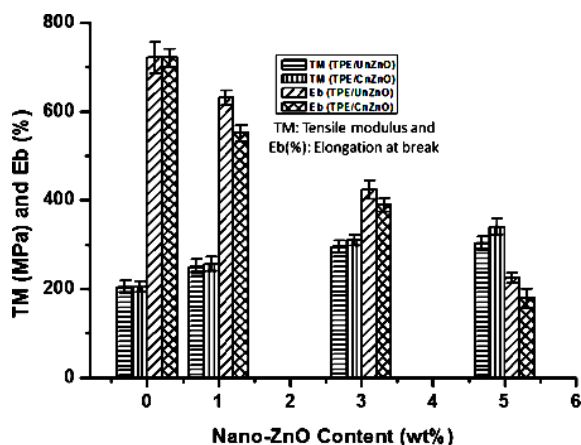


Figure 5. Tensile and elongation at break of TPE/nZnO nanocomposites.

Electrical Properties

The dielectric constant and resistivity of both uncoated and coated TPE/nZnO nanocomposites were examined with nZnO content at a constant frequency of 10 kHz. Figure 6(a) displays the variation of the dielectric constant of uncoated and coated TPE/nZnO nanocomposite against nZnO content. The dielectric constant of nanocomposites increases virtually linearly with increasing nZnO content. There

is no sudden increase in the dielectric constant in the nZnO content, which is known as the percolation phenomenon, also occurs for these nanocomposites when the nZnO content is studied at about 5 wt%. This is probably due to the low dielectric property of nZnO. This behavior is very different from that observed in other polymers/high dielectric fillers (such as carbon nanotubes) in nanocomposites, where percolation thresholds or concentrations exist [26]. Cluster structures are formed in most polymer/filler composites because of the agglomeration of filler particles at percolation thresholds. Such a structure is suitable for the formation of filler cluster in relatively low filler content. From this, it is clear that the fillers of TPE/nZnO nanocomposites are dispersed as isolated particles in the matrix instead of the agglomeration of clusters with low nZnO content. The dispersion of fillers in the polymer matrix is another important factor that influences the dielectric behavior of the composite. SEBSMA is usually an effective compatibilizer for dispersing fillers in polyolefin such as polypropylene or polythene [27]. In this respect, the addition of SEBSMA in TPE/nZnO nanocomposite will increase the distribution of ZnO nanoparticles in the TPE matrix, thus inhibiting ZnO nanoparticles from aggregating and creating a path network.

Figure 6 (b) shows resistivity to the nZnO content of TPE/nZnO nanocomposites. The resistivity of virgin TPE is $\sim 2.6 \times 10^{11} \Omega\cdot\text{m}$. The resistivity of TPE/nZnO nanocomposites begins to decline up to 2 wt% with the addition of nZnO content, then it decreases almost continuously by increasing the nZnO content. The resistivity of nanocomposites in 5 wt% nZnO decreases to $\sim 0.24 \times 10^{11} \Omega\cdot\text{m}$. This corresponds to about 91 reductions for virgin TPE. Resistivity in this respect can be described by percolation theory.

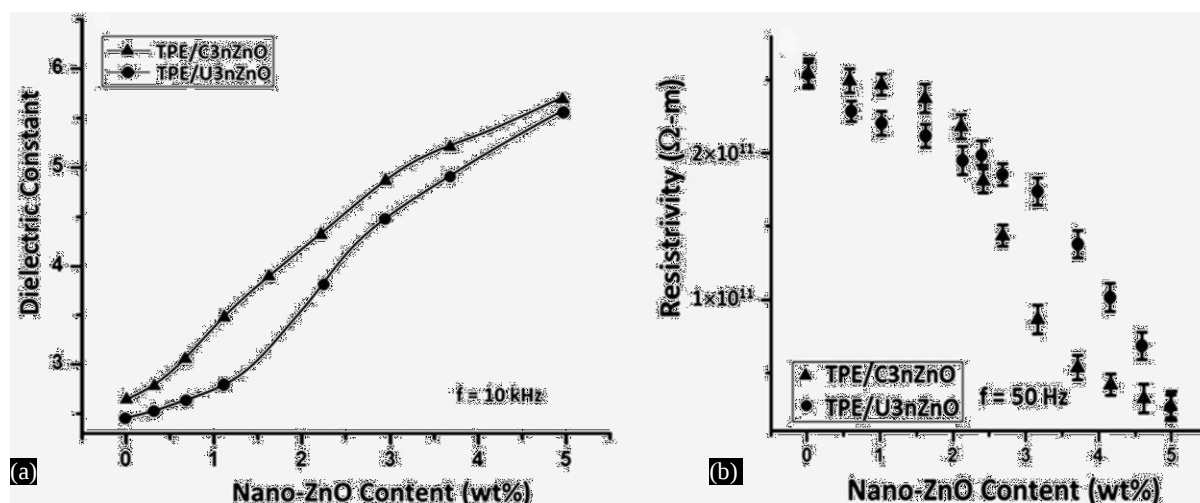


Figure 6. (a–b) (a) Dielectric constant and (b) Resistivity of TPE/nZnO nanocomposites as a function of nano-ZnO content (wt%).

CONCLUSION

A melt mixing process has been prepared using PBT-PTMG based TPE/nZnO nanocomposite, which will be followed by the hot compression molding method. This work examined the effects of surface-modified nZnO particles on the mechanical, morphological, thermal, and electrical properties of the TPE matrix nanocomposites. Adding nZnO to polymers has improved its rigidity, strength, and thermal stability but has reduced dramatically elongation at break. As a result, morphological studies have shown that there is better interaction between the SEBSMA coated filler and the matrix at 3 wt% filler concentration. The addition of SEBSMA coated-nZnO has improved the thermal properties of the composites due to the small and uniform crystal size distribution. The effect of coating-nZnO particle size on dielectric constant and resistivity properties has been investigated. The outcomes show that the dielectric constant of nanocomposites increases nearly linearly with increasing nZnO concentration. When the nZnO content goes above 2.5 wt%, the resistance of the nanocomposites decreases sharply.

This feature can be described by percolation theory. It is concluded that the addition of fillers showed improved mechanical, thermal as well as electrical properties.

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